Aerosol Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/uast20

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Published online: 20 Oct 2011.

To cite this article: Subhasis Biswas , Philip M. Fine , Michael D. Geller , Susanne V. Hering & Constantinos Sioutas (2005) Performance Evaluation of a Recently Developed Water-Based Condensation Particle Counter, Aerosol Science and Technology, 39:5, 419-427, DOI: 10.1080/027868290953173

To link to this article: http://dx.doi.org/10.1080/027868290953173

Aerosol Science and Technology, 39:419–427, 2005

ISSN: 0278-6826 print / 1521-7388 online
DOI: 10.1080/027868290953173

Performance Evaluation of a Recently Developed Water-Based Condensation Particle Counter

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This study provides an intercomparison of the performance of a newly developed water-based condensation particle counter (W-CPC) and a more widely used butanol-based CPC (TSI 3022A). Four test aerosols (ammonium nitrate, ammonium sulfate, adipic acid, and glutaric acid) were generated and tested in the laboratory before the instruments were deployed at four field locations (USC/downtown LA, I-710 Freeway, Pacific coast, and Los Angeles International Airport). Both instruments sampled the same incoming aerosol. Selected experiments utilized a differential mobility analyzer to select a particle size upstream of the CPCs. Evaluation of performance was based on the response of the instruments to varying particle composition, concentrations, and size. The results indicated good correlation between the two CPCs, with R^2 values ranging from 0.74–0.99. Good agreement was found between the two instruments for particle concentrations between 0 and 40,000 particles/cm^3, with W-CPC/TSI 3022A ratios between 0.8 and 1.2. Due to differences in the photometric mode calibration of these instruments, the ratio drops to 0.6–0.8 between 40,000–100,000 particles/cm^3. However, the ratio rises again for lab aerosols above 100,000 particles/cm^3 to 1.0–1.1. Results of this evaluation show that the W-CPC is a reliable particle-counting technology for particle concentrations encountered downstream of a DMA as well as in some ambient environments ($<$40,000 particles/cm^3).

INTRODUCTION

Numerous epidemiological and toxicological studies have demonstrated significant associations between health effects and airborne particulate matter (PM). For instance, cohort studies have showed increased mortality with long-term exposure to fine PM and particulate sulfates (Dockery et al. 1993; Pope et al. 2002). While most studies focused on PM mass (Pope 1991; Hoek and Brunekeef 1993), recent findings suggest that particle number concentration, rather than mass, may be an alternate predictor of health effects (Laden et al. 2000; Oberdorster et al. 1990; Pekkanen et al. 1997; Peters et al. 1997). Ambient particle number concentrations are dominated by smaller particles in the ultrafine size range (diameters less than 100 nm), and there is growing evidence of increased chronic and acute health effects due to exposure to ultrafine particles (Oberdörster and Utell 2002; Ibald-Mulli et al. 2002; Li et al. 2003, 2004; Xia et al. 2004). The capability of individual particles to penetrate cellular membranes and cause damage has also been demonstrated (Li et al. 2003). Ultrafine particles are generated by gas-to-particle conversion during combustion processes (in the combustion zone, near stack or near tailpipe) or via secondary formation in the atmosphere (Shi et al. 1999; Hitchins et al. 2000, Zhang et al. 2004). Since particle number concentrations and particle size distributions are important metrics of both atmospheric processes and human exposure to PM, accurate measurements of these parameters are essential.

Several continuous monitors such as condensation particles counters (CPCs) or condensation nuclei counters (CNCs) are available to provide real-time number concentration measurements of airborne particles. CPCs are also used in conjunction with differential mobility analyzers (DMAs) to determine particle size distributions (Woo et al. 2001). CPCs optically count particles that are enlarged by a condensation of vapor. Growth of particles is achieved when a particle-laden air stream is subjected to supersaturation of a condensable vapor that quickly condenses onto the particle surfaces, resulting in particle growth. CPCs can achieve supersaturation by adiabatic expansion and cooling, thermal and molecular diffusion, and rapid mixing (Hämeri et al. 2002). The lower particle size cutoff, i.e., the smallest particle that is activated for condensational growth, depends on the characteristics of the particles and vapor, the degree of the

Received 1 December 2004; accepted 8 March 2005.
The authors thank Fred Quant at Quant Technologies for the temporary loan of the W-CPC for testing. This work was supported by the Southern California Particle Center and Supersite (SCPCS) funded by the USEPA (STAR award #R82735201). This manuscript has not been subjected to the EPA peer and policy review and therefore does not necessarily reflect the views of the Agency. No official endorsement should be inferred.
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supersaturation, the physical configuration of the growth area, and the sampling pressures, temperatures, and flow rates (Mertes et al. 1995; Liu and Kim 1977; Wilson et al. 1983; Bartz et al. 1985; Wiedensohler et al. 1997; Hermann and Wiedensohler 2001; Zhang and Liu 1991; Ankilov et al. 2002).

Several studies have been carried out to evaluate the performance of CPC technology via intercomparison of two or more CPC types. Wiedensohler et al. (1997), for example, determined the particle detection efficiency curves and 50% particle detection efficiency diameters for commercially available continuous-flow CPCs (CPC TSI-3760, CPC TSI-3010, and UCPC TSI-3025, TSI Inc., Shoreview, MN, USA) and calibrated twenty four particle counters for different operating conditions (different flow rates and temperature differences between saturator and condenser). It was found that the integral counting efficiency decreases at reduced pressure and increases with increased temperature difference between saturator and condenser (Kim et al. 2002a; Zhang and Liu 1991; Hermann and Wiedensohler 2001). Sem (2002) reviewed the performance of the TSI 3010, 3022A, and 3025A CPCs, while Matson et al. (2004) evaluated a TSI CPC 3007 and a TSI P-Trak™. The TSI 3025A CPC has been found to count approximately 10% more particles than the TSI 3022A (Harrison et al. 1999), presumably due to a lower particle activation size. When large and rapid variations in aerosol number concentrations are to be measured, the response time of the CPC becomes an important factor. The TSI CPC 3025 has a faster time response than CPC TSI 3010 and thus is capable of better time resolution (Buzorius 2001).

Aerosol Dynamics Inc., in partnership with Quant Technologies and TSI, Inc., recently developed a new continuous, laminar flow, water-based CPC (W-CPC) (Quant Technologies WCPC-400, equivalent to TSI, Inc. model 3785; Hering and Stolzenburg 2004; Hering et al. 2005). It has a lower operating cost, avoids the toxicity and nuisance of alcohol fumes and spills (a feature that is particularly important when using these instruments indoors), and promises better time response than the alcohol-based CPCs. The purpose of this study is to examine the performance of this W-CPC in comparison with the commercially available TSI 3022A CPC. The study presents the comparative response of these instruments as a function of particle size, particle composition, and number concentration for both laboratorygenerated and ambient aerosols. Since the TSI 3022A is currently in wide use, it was taken as the standard to which the new W-CPC was compared.

METHODS

Instruments

Two continuous CPCs, a TSI 3022A and the W-CPC (Quant Technologies, WCPC 400) were compared in this study. The TSI 3022A CPC uses n-butyl alcohol as the working fluid. It has two inlet flow modes, a high flow (1.5 lpm) and a low flow (0.3 lpm), which give it operational flexibility. The 50% lower particle size detection limit, as specified by the manufacturer, is 7 nm. The 3022A CPC uses three modes of particle counting: single-particle real-time counting ($<$1,000 particles/cm^3), single-particle live-time counting (1,000–10,000 particles/cm^3), and photometric mode ($>$10,000 particles/cm^3; Sem 2002). For real-time counting the effective sample time is equated to the actual sample time, while for live time counting the effective sample time is set equal to the actual sample time minus the cumulative time the light signal is larger than the detection threshold. At higher concentrations multiple particles are present in the scattering volume, and the particle concentration is derived from the light scattered from the cloud of particles within the scattering volume (photometric mode). The photometric mode of the 3022A CPC used in this study was recalibrated by the manufacturer (TSI Inc.) within two months of our measurements. For both the 3022A and the W-CPC, the calibration of the photometric response is done using DMA-classified 50 nm NaCl aerosol. According to Sem (2002), the 3022A has a response time between about 7 and 10 s.

The W-CPC is a laminar-flow, thermally diffusive instrument, as is the alcohol-based CPC. Instead of a condenser, the W-CPC has a "growth tube" that permits the use of water as the working fluid, as described by Hering and Stolzenburg (2004; Hering et al. 2005). The walls of the condensing region of the growth tube are warmed and saturated with water. Since the mass diffusivity of water vapor is higher than the thermal diffusivity of air, the water vapor flux to the centerline of the tube is faster than the heat flux from the walls, creating supersaturation along the centerline of the tube. Once enlarged, particles are detected optically. It is an unsheathed instrument with an operating flow rate of 1.0 l/min. For particle concentrations below approximately 30,000/cm^3, number concentrations are derived through single-particle counting with a correction for the deadtime. For the W-CPC the dead time is estimated as the cumulative time the light signal is larger than the detection threshold, multiplied by an empirically determined dead-time correction factor. This factor accounts for the overlap of tails of adjacent pulses and is determined for each instrument using a NaCl test aerosol, as described in Hering et al. (2005). At higher concentrations the instrument operates in photometric mode, similar to the TSI-3022, and concentrations are based on empirical calibration of the total light scattering from the particle cloud within the viewing volume. The response time of the W-CPC, including transport lag, is 1.3 s, significantly faster than the 3022A (Hering et al. 2005). Further details are given in Table 1.

Experimental Design

Several indoor and outdoor experiments were conducted to examine particle-size-dependent performance, effects of particle composition, and biases related to absolute total number concentration. The size-dependent response of the W-CPC and TSI 3022A were carried out by connecting a differential mobility analyzer (DMA, from a scanning mobility particle sizer, SMPS 3080, TSI Inc.) to a common inlet of these two instruments as

TABLE 1
Characteristics of the two CPCs according to TSI, Inc. user manuals and literature

	TSI 3022A	W-CPC
Particle size range		
Minimum detectable particle (D_{50})	7 nm	5 nm
Maximum detectable particle	>3 μm	>3 μm
Particle concentration		
Single-count mode	0–1,000 particles/cm^3 (real time)	0 to $\sim$30,000 particles/cm^3 (dead-time corrected)
	1,000–10,000 particles/cm^3 (live time)	
Photometric mode	10^4 to 10^7 particles/cm^3	$\sim$30,000 to 10^7 particles/cm^3
Particle concentration accuracy	$\pm$10% at $<5 \times 10^5$ particles/cm^3	$\pm$10% at $<3 \times 10^4$ particles/cm^3
	$\pm$20% from 5×10^5 to 9.99×10^6	Not specified $>3 \times 10^4$ particles/cm^3
False background counts	0.01/cm^3	$<$0.001 particle/cm^3 1 h average
Response time	$<$13 s for 95% response to concentration step change	$<$2 s for 95% response to concentration step change

shown in the Figure 1. The SMPS was equipped with a 0.71 μm impactor inlet. The common inlet was made of a T-junction, and proper care was taken so that the two CPCs were connected with same short length (8 cm) of conductive tubing to minimize diffusional losses. The aerosol inlet flow rate for the W-CPC and the 3022A were 1.0 and 0.3 lpm, respectively. The sheath flow-rate for the SMPS was set to 13.0 lpm, giving a 1:10 ratio of aerosol to sheath flow. The range of particles selectable under these flow conditions was between 6.5 and 265 nm. Various sizes of monodisperse particles (10, 20, 30, 40, 50, 60, 70, 80, 90, 100, 120, 150, 180, 220, and 260 nm in mobility diameter) were selected from a polydisperse source by the SMPS, and particle counts were recorded either visually from the CPC screens, or by two laptop computers connected to analog outputs of the two CPCs. The sampling periods were generally between 1 and 2 min for each particle size. For the 3022A, Aerosol Instrument Manager software (version 4.0 TSI inc, St. Paul, MN, USA) was used, while the W-CPC employed custom software. One-second time resolution data was stored. To measure the total number concentration of particles, the common inlet of the CPCs was separated manually from the SMPS and connected to either the outlet of the aerosol generation unit or exposed to ambient air.

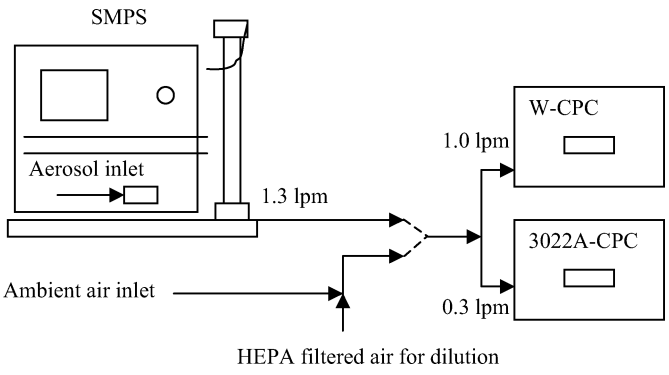

FIG. 1. Experimental setup. Dotted lines indicate alternate inlet configurations, the upper inlet used for size-dependent response measurements, and the lower inlet attached for total number concentration measurements.

Laboratory-Generated Particles

The performance of the W-CPC was examined with various types of polydisperse laboratory-generated aerosols. Ammonium sulfate, ammonium nitrate, glutaric acid, adipic acid, and 60 nm polystyrene latex (PSL) particles were generated by atomizing their respective solutions with a constant output Nebulizer (HEART, VORTRAN Medical Technology, Inc., Sacramento, CA, USA). Ammonium sulfate and ammonium nitrate are the two most predominant inorganic salts in particulate matter in the U.S., while adipic and glutaric acids are known secondary organic aerosol components found in ambient PM (Cruz and Pandis 1999; Sempere and Kawamura 1994). The solution containing 60 nm PSL particles also contains surfactant additives to avoid particle coagulation. This results in the production of some polydisperse surfactant-only residual particles in addition to PSL particles. In any case, both PSL and the surfactant are hydrophobic, and these tests aimed to examine the effects of hydrophobicity on instrument response. The generated particles were mixed with sufficient dilution of room dry air in a 2 l glass container to remove excess moisture. Particle concentrations in the room air were insignificant in comparison to the concentrations of generated particles. The dry aerosols were then passed through a series of Po-210 neutralizers (NDR Inc., Grand Island, NY, USA) to neutralize excess particle charge and bring the charge distribution to Boltzmann equilibrium conditions. Total number counts, as well as size-selective experiments, were

performed by attaching the common inlet of the CPCs or the SMPS to the particle generation unit. Total number concentration levels were varied via dilution with HEPA filtered air.

Field Evaluation

The field evaluation of the W-CPC was carried out by placing all of the instruments in a sampling van and parking it at different locations in Southern California that are influenced by different particle sources. The University of Southern California's Particle Instrumentation Unit of the Southern California Supersite (USC) is located 2 miles south of downtown Los Angeles and about 100 m downwind of a major freeway (I-110). It represents a typical urban/traffic/industrial environment. Los Angeles International Airport (LAX) was selected because it is one of the busiest airports in the world with 60,000 aircraft operations (takeoff and landings) per month (Yu et al. 2004) and creates very high and rapidly fluctuating number concentrations downwind of the runways. The sampling was conducted about a half mile downwind of the southern runway where flights were landing. To test coastal background nonurban aerosols, sampling was carried out at a beach on the Pacific coast, upwind of LAX. The final sampling location was adjacent to the I-710 freeway in Downey, CA. The I-710 freeway has eight lanes and is a major truck route in Southern California between the port areas and the warehouses and rail yards east of downtown Los Angeles. More than 25% of the vehicles are heavy-duty diesel trucks in the middle of the day when the sampling was conducted (Zhu et al. 2002), and thus particles at this location are characterized by a relatively high elemental carbon content. The field evaluations included total number concentration comparisons at all of the locations. Each of these sampling periods lasted approximately 20 mins, and number concentrations were recorded at 1 s intervals. The exception was USC, where the total sampling period was about 10 h, and 1 min average data was recorded by the software. Also at USC, the W-CPC was run concurrently with two 3022A instruments, and the three CPCs were connected to a common inlet. The two collocated 3022As provided a measure of precision of these instruments. Our results showed that the 3022A has a high degree of precision ($R^2 = 0.99$, average ratio of 1.06 ± 0.05). Size-selected monodisperse particle tests were conducted at both USC and the I-710 freeway. At the I-710 location, the intake flow of the CPCs was diluted to explore the response of the CPCs at different absolute number concentrations of the same aerosol, thus providing a wider range of absolute number concentrations for testing. Similar to the laboratory tests, a HEPA filter and a needle valve were connected via a tee to the common inlet, and the valve adjusted to give different levels of dilution and thus number concentration.

RESULTS AND DISCUSSION

Previous studies have demonstrated that particle size can have a significant impact on the performance of CPCs, especially near the lower size detection limit. This size is of particular significance, as small deviations in the cutoff size may result in substantial differences in total number concentration readings among CPCs due to the general increase in particle number concentration with decreasing particle size of ambient aerosols. This is even more pronounced in situations characterized by high ambient particle counts and small number modes, such as nucleation phenomena (Zhang et al. 2004; Kulmala et al. 2004) or the immediate environments of roadways (Kittelson et al. 2004; Zhu et al. 2002). In this work, we have selected particles between 10 and 260 nm by a DMA (TSI 3080) for four laboratory-generated aerosol compositions and ambient aerosols at two locations (USC and I-710). Figure 2 displays the effects of particle size on CPC response. The ratio of the W-CPC response to the 3022A response ranges from 1.0 to 1.1

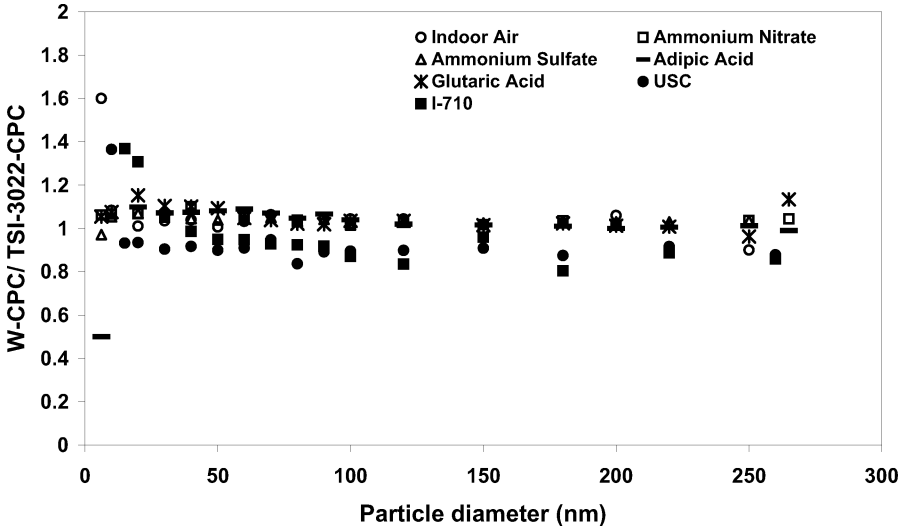

FIG. 2. Size-dependent performance of the W-CPC versus the TSI 3022A.

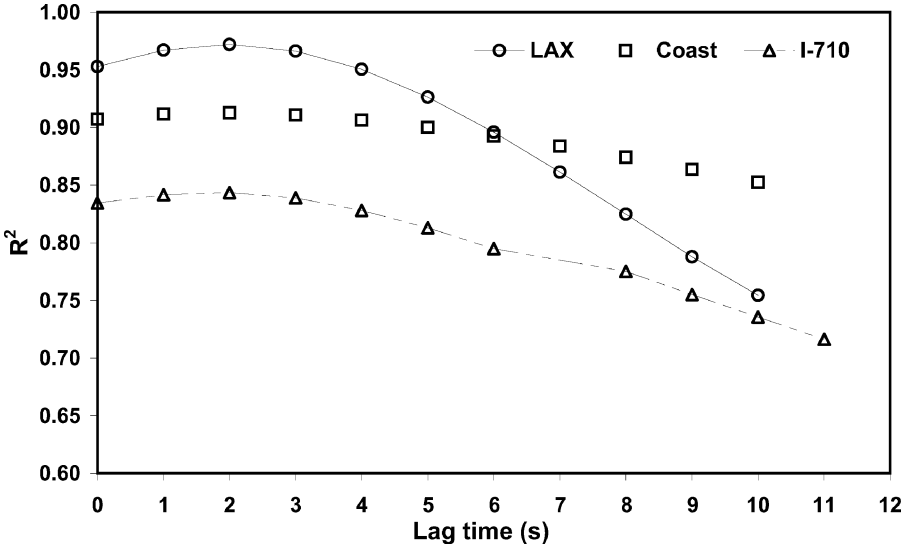

FIG. 3. R^2 (Pearson) versus lag time between the W-CPC and the TSI 3022A (TSI 3022A lagging behind the W-CPC).

for the smaller laboratory-generated particles (10–50 nm). For the smallest sizes (less than 15 nm), the slightly higher concentrations measured by the W-CPC may be due to the lower particle diameter detection limit of the instrument. Although the particle size distributions are theoretically monodisperse, they actually consist of a distribution of particles, some of which may occur below the lower size detection limit of the 3022A. As the laboratory-generated particles increase in size, the ratio approaches 1.0. Note that the results are generally similar for all particle types tested, indicating that particle chemical composition did not have a significant effect on relative instrument response based on the types of laboratory aerosols tested. The W-CPC indicates lower concentrations than the TSI-3022 for the ambient aerosols at USC and the I-710 Freeway, with ratios generally ranging from 0.8 to 1.0. A paired *t* test (two-tail) between the average response ratio of all lab aerosols and that of the I-710 and USC resulted in *p* values of 0.012 and 0.005, respectively. Therefore, there is a significant difference between the mean ratio for laboratory and for ambient aerosols across the entire particle size range.

In these urban environments, however, the W-CPC to the 3022A ratio seems to be increasing with decreasing particle size, particularly looking at the data obtained at the I-710. Overall, the agreement is quite good and within the manufacturer-specified ranges of uncertainty for each device (±10% for each). For this size-selective testing, the number concentrations were well below 10,000 particles/cm^3, so both instruments were operating in single-particle counting mode.

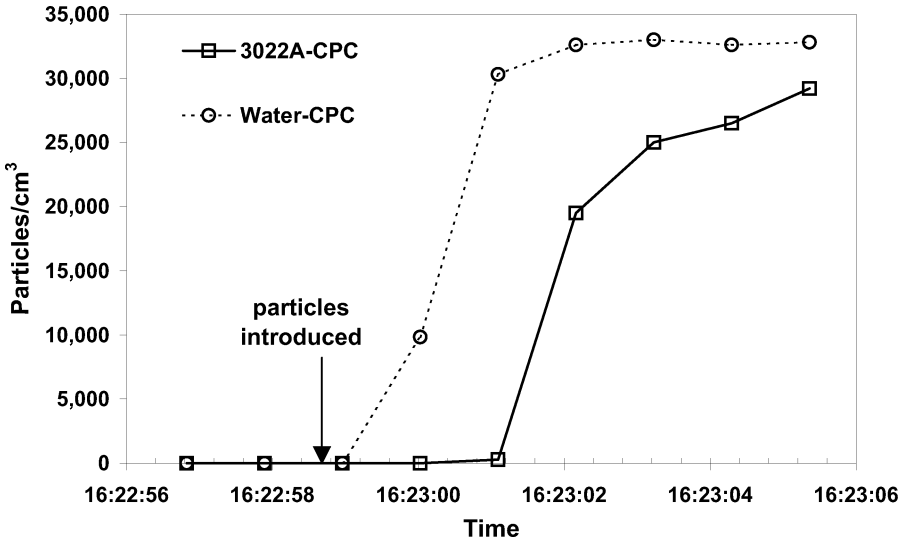

FIG. 4. Time response of CPCs upon introduction of particles.

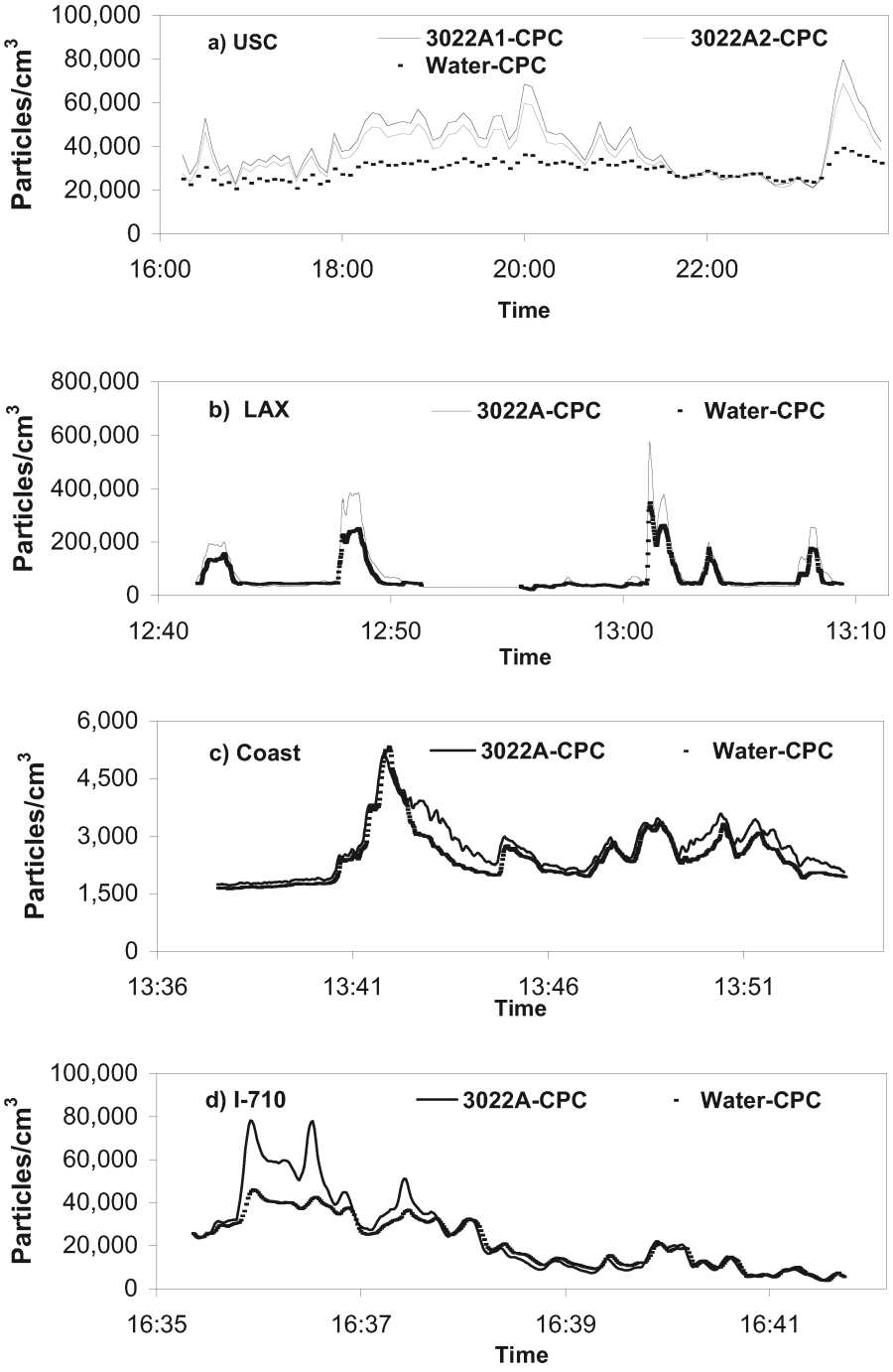

FIG. 5. Time series response of the CPCs at different ambient locations.

Next, the CPCs were operated without a DMA upstream in order to measure total particle concentrations of both ambient and laboratory-generated particles. It became immediately clear that the second-by-second response of the two CPCs were offset by a relative lag time, presumably due to differences in flow rates and internal plumbing configurations. The actual lag time was determined by finding the time lag producing the maximum correlation between the concentration readings of the two instruments. Figure 3 shows how the correlation between the two CPCs reading ambient aerosol changes with different lag times. The figure shows that a 2–3 s lag time brings the instruments into the best agreement, and thus a 3 s lag time was applied to all concurrently collected data. This result is further supported by Figure 4, which shows the response of the instruments when particles were first introduced after removal of a HEPA filter from their common inlet. The 3022A is again seen to lag the

W-CPC by approximately 2–3 s. Also note that the W-CPC responds very quickly and reaches near steady state values within 1–2 s of the introduction of particles.

Selected time series of total particle number concentrations at different outdoor locations, smoothed by a 5 s moving average and adjusted for lag time, are shown in Figure 5. At USC, 5 min average data is displayed over a longer sampling period. The instruments generally track each other well, but there are some noticeable biases. The site at USC exhibited a particle number concentration ranging between 16,000–75,000 particles/cm^3, which is typical of urban environments with traffic influences (Kim et al. 2002b; Sardar et al. 2004). The W-CPC reads approximately 20% fewer particles on average throughout the period of sampling, an observation in line with the results shown in Figure 2 showing the W-CPC reading slightly lower concentrations across different sizes for particles above 50 nm.

Data from LAX displays rapid fluctuations in particle concentrations (18,000–600,000 particles/cm^3) with the peaks corresponding to aircraft landings. It is of particular note that these concentration peaks far exceed the concentrations measured immediately adjacent to the I-710 freeway, which is noted for its heavy-duty diesel truck traffic that generally account for over 25% of the total number of vehicles in that freeway (Zhu et al. 2002). The W-CPC is lower than the TSI-3022 at these high concentrations, something that was also observed in the I-710 freeway results, depicted in Figure 5d. The last 5 min of the I-710 data in Figure 5d corresponds to sampling during filtered-air dilution at several ratios in order to sample over a wider range of number concentrations (5,000–100,000 particles/cm^3). All subsequent analyses from this site includes these data. The coastal sampling showed very low background concentrations of 1,700–5,200 particles/cm^3 and generally excellent agreement between the two CPCs.

TABLE 2
Statistical comparisons between the two CPCs

	R^2	Average Ratio (±) SD	Range of Concentrations (particles/cm^3)
Field locations			
LAX	0.96	0.94 ± 0.27	18,000–600,000
I-710 freeway	0.91	0.91 ± 0.23	5,000–100,000*
USC	0.74	0.80 ± 0.15	16,000–75,000
Coast	0.91	0.94 ± 0.13	1,700–5,200
Laboratory aerosols			
Ammonium sulfate	0.99	0.92 ± 0.22	20,000–1,000,000
Ammonium nitrate	0.99	1.0 ± 0.34	15,000–600,000
Adipic acid	0.97	0.88 ± 0.4	20,000–1,000,000
Glutaric acid	0.94	0.88 ± 0.25	5,000–1,500,000
60 nm PSL with surfactant	0.86	1.0 ± 0.20	30,000–125,000

*Includes filtered-air dilution sampling.

Table 2 summarizes the statistical comparisons between the two CPCs measuring total number counts at the ambient sites as well as the laboratory-generated aerosols. R^2 values over the ambient sampling periods (depicted in Figure 5) ranged from 0.74 to 0.99 after adjusting for the lag time. The correlation coefficients are fairly high in all the cases, and the W-CPC/3022A ratio varied between 0.80 and 1.0. The ratio was lowest for USC and highest for ammonium nitrate particles in the lab. As will be discussed below, these differences are most affected by the absolute number concentrations and the CPC counting mode rather than differences in particle source or characteristics.

Figure 6 shows the concentration-dependent performance of the W-CPC, again using the ratio of total number concentration

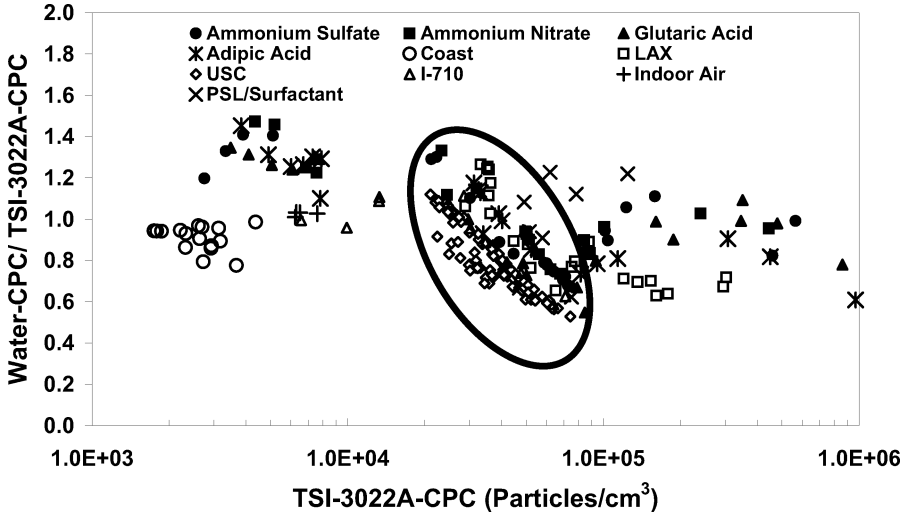

FIG. 6. Total number concentration dependent performance of the W-CPC versus the TSI 3022A. Oval indicates size ranges with systematic bias between the instruments.

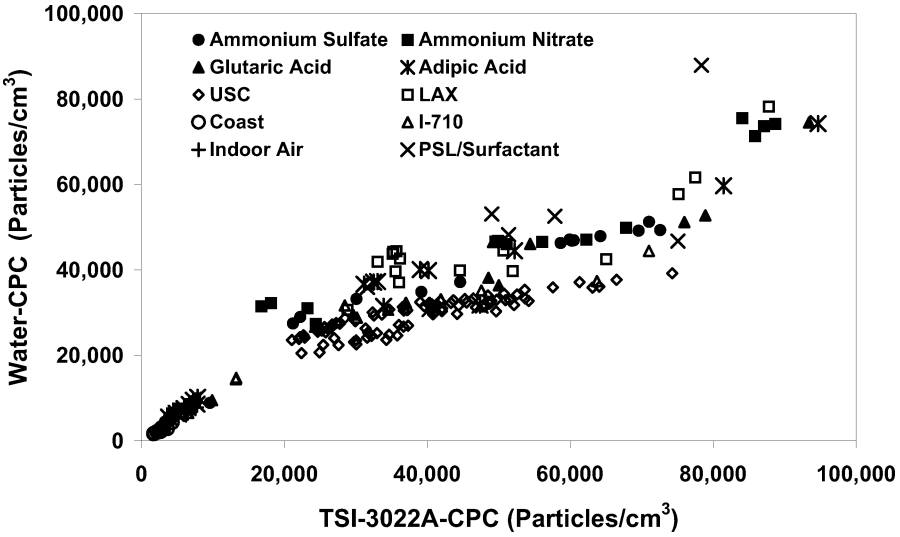

FIG. 7. Correlation between the CPCs in the photometric mode transition range.

of the W-CPC to the 3022A that was used as a reference. The figure includes 1 min average data for clarity, except for USC, where the data is averaged over 5 min. The W-CPC generally reads higher particle numbers than the 3022A below 30,000 particles/cm^3, except at the coastal and USC sites, where ratios are between 0.8 and 1.0. The W-CPC/3022A ratio generally decreases, irrespective of particle origin, between 30,000 and 80,000 particles/cm^3, as shown in the encircled segment of the figure. This is within the range of concentrations at which both CPCs are either operating in or switching to the photometric counting mode. Figure 7 also demonstrates this effect, where the W-CPC reads lower particle concentrations than the 3022A between about 30,000 and 80,000 particles/cm^3. There is good correspondence between the W-CPC and the 3022A ($R^2 = 0.89$, slope = 0.94) below 30,000 particles/cm^3. The transition region where the W-CPC concentrations are lower may be due to an issue with the photometric calibration. Both Figures 6 and 7 show that at about 80,000 particles/cm^3 the W-CPC resumes reading increases in concentrations at a similar rate as the 3022A, and the ratio increases to near 1.0 for most laboratory-generated particles when number concentrations exceed 100,000/cm^3. However, in the case of LAX, where there are large spikes in number concentrations arising from aircraft emissions, the W-CPC reads lower particle numbers than the 3022A with a ratio near 0.7–0.8.

The particles corresponding to peak concentrations at LAX originating from aircraft engine emissions are small and rich in more hydrophobic material, such as organic and particularly elemental carbon (Tesseraux 2004; Johnson et al. 2003). For hydrophobic particles with diameters well above the cutpoint, the W-CPC exhibits high counting efficiencies at high number concentrations, as indicated by agreement reported here for 60 nm polystyrene latex particles mixed with its surfactant (Table 2, Figures 6 and 7). Additionally, Hering et al. (2005) report high counting efficiency for concentrations as high as 90,000 particles/cm^3 for 80 nm oleic acid, a hydrophobic aerosol. However, for the measurements at LAX it is likely that a significant fraction of the particles are near the lower detection limit for both counters. At the high concentrations generated by the aircraft plume both CPCs will be affected by depletion of the condensing vapor. This vapor depletion will raise the lower detection particle size of both instruments. At low concentrations the lower cut size is very similar between the 3022 and the W-CPC for hydrophobic as well as hydrophilic particles. But, as a function of concentration the cut size for the W-CPC may well increase faster than that for the 3022, causing the W-CPC to report lower concentrations.

SUMMARY AND CONCLUSIONS

The W-CPC showed similar response to the 3022A as a function of particle type and particle size within the stated uncertainties of the manufacturers (±10%). In general, neither the hygroscopicity nor the diameter of the particles tested resulted in significant biases. The only factor significantly affecting the relative performance of the two instruments was the absolute particle number concentration. The W-CPC tended to report lower number concentrations than the 3022A in the range from 30,000 to 100,000 particles/cm^3. This consistent observation may be due to differences in the photometric calibrations of the two instruments. This issue requires further investigation and may be correctable with new calibrations and programming. For number concentrations below 30,000 particles/cm^3, the relative agreement was good, indicating that the W-CPC is accurate for cleaner environments and for particle-sizing applications with a differential mobility analyzer deployed upstream.

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